



A bath of crystal-clear molten salt helps produce strategic nuclear materials and effectively transforms nuclear power into a safe, renewable energy source.

NUCLEAR ENERGY AS A RENEWABLE?

"It's not as crazy as it sounds," says Marisa Monreal, an actinide chemist at Los Alamos. "Conventional nuclear power consumes refined uranium and produces radioactive waste. Definitely not renewable. But a breeder reactor can produce more nuclear fuel than it consumes, and new designs offer extraordinary improvements in safety and energy output, with the ability to consume some radioactive waste. With fuel inputs and waste outputs so low, it comes very close to being a renewable."

Matt Jackson, a close colleague of Monreal's, is equally optimistic. "The fuel-use efficiency could be hundreds of times that of nuclear plants today," he says. "When very little fuel is wasted, very little fuel is needed."

With the right reactor design, some experts have argued, nuclear energy will deserve a place among the more widely recognized renewables, which also have limitations to their renewability. Biofuel, for example, consumes various resources, such as fertilizer, and produces waste, such as soot. Geothermal energy draws power from the heat found deep underground—heat mostly generated by natural radioactivity, a form of nuclear energy—and dredges up some toxic material in the process.

Monreal and Jackson expect that the right reactor design will see low-grade nuclear fuel immersed in a molten salt—essentially a liquefied rock that flows like water. It's just a matter of studying the bizarre stuff.

#### Lava laboratory

Molten salts are valuable to many nuclear technologies. Monreal and Jackson currently pursue two of these applications—power production and plutonium processing (both discussed herein)—although there are several others, such as nuclear safeguards, uranium purification, and energy storage, that can benefit from molten-salt development as well. For the sheer scope of its potential benefit, however, power production is perhaps the most tantalizing.

Monreal, Jackson, and other next-generation nuclear-power pioneers imagine eliminating the contents of today's reactors—enriched fuel rods wrapped in engineered cladding and cooled with a mechanically managed flow of pressurized water, all of which are simply entombed and replaced once spent—and instead

**Electron flow** 400 Electrical current source ANODE (+) CATHODE (-) **MOLTEN SALT** CaCl<sub>2</sub> Plutonium oxide (PuO<sub>2</sub>) <u>Plutonium</u> AT THE CATHODE:

2Ca + PuO<sub>2</sub> → 2CaO + Pu metal

filling a reactor with a single liquid: a molten salt, with a much more easily produced nuclear fuel simply dissolved in.

Any number of salt compounds might do the trick. One that has been experimented on extensively, for example, is a mixture of lithium and beryllium fluorides (written as FLiBe). But it can also be something more familiar, such as sodium chloride, which is ordinary table salt, or calcium chloride, another common salt. That these compounds are known as "salts" refers to their construction from positive ions (generally from the first two columns of the periodic table, like lithium or sodium) electrostatically paired with negative ions (from the second-to-last column, like fluorine or chlorine).

A mixture of chloride salts is currently loaded in an electrochemical cell in a deceptively nondescript laboratory at Los Alamos. It has been heated to more than 800 degrees Celsius, nearly 1500 Fahrenheit, causing it to melt to a liquid. But far from the thick, viscous lava one might expect from molten rock, the salt stirs easily.

"It has remarkable chemical and thermophysical properties for nuclear applications," says Jackson. "High density for desirable neutronics, low viscosity for easy pumping, lower corrosivity (compared to fluorides) for practical containment, good conductivity for catalytic processing, and high solubility for dissolving important actinide compounds into it, such as uranium or plutonium chloride."

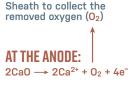
The concept of a molten-salt reactor (MSR) itself is not new, although the push toward using an advanced fuel-salt composition is. Throughout the 1960s, Oak Ridge National Laboratory built and experimented with a small MSR. Their experiments were quite successful; the reactor worked safely and reliably, fission

products and other contaminants that accumulated in the salt were generally unproblematic, and key parameters were measured that either confirmed or improved upon previous theoretical calculations. In fact, the whole enterprise seemed distinctly promising, but unfortunately, that promise came just a little too late. By the late 60s and early 70s, funding and infrastructure were already well committed to the entrenched industry of enriched solidfuel, water-based reactors.

Today, however, interest has been renewed with proposals for potentially revolutionary improvements in both major components of the MSR: the fuel and the salt.

#### **End of enrichment**

The primary fuel tested at Oak Ridge was based on uranium-235 (U-235), which also powers most of today's commercial reactors. It comprises less than 1 percent of uranium found in nature, so it must be obtained through enrichment: separating it from the much more abundant U-238 isotope,



electrochemical cell at Los Alamos, researchers have successfully converted metal oxides into metals. Success with tin and cerium oxides has been demonstrated so far, with the ultimate objective of obtaining plutonium metal from plutonium oxide as a way to help meet increasing weaponsproduction demands.

In an experimental

2Ca<sup>2+</sup> + 4e<sup>-</sup> → 2Ca

which itself cannot produce a sustained chain reaction to power a reactor. But enrichment is a cumbersome, costly, and wasteful process. Enriching naturally occurring uranium to just 3–4 percent U-235, which is adequate for power production, consumes a great deal of energy and typically results in less than 10 percent of the unenriched starting material becoming enriched to that level. The remainder, called "depleted uranium," gets repurposed for non-fission applications, even though it still contains a good 40 percent as much U-235 as naturally occurring uranium; this resource goes unused.

"Enrichment has been widely viewed as a necessary evil, both in today's commercial reactors and in earlier experimental ones," Jackson says.

However, just because the Oak Ridge experiment used enriched fuel doesn't mean an MSR has to. An MSR is particularly amenable to being built as a breeder reactor, which breeds its own fuel. Instead of neutrons being used to split U-235 nuclei, it can be set up for neutrons to merge with U-238. The resulting U-239 decays in short order to plutonium-239 (Pu-239), which is an excellent fission fuel, on par with U-235. The entire enrichment process can be skipped.

One need only create a uranium compound that dissolves well in the liquid salt. With a fluoride-based salt, uranium fluoride works best; likewise, for a chloride salt, it's uranium chloride. Either way, the process is not a full-blown nuclear separation but rather a comparatively simple chemical conversion, and one that is performed regularly for uranium research applications anyway. And since the uranium fuel is just dissolved into the liquid, it can remain there until converted to Pu-239 and brought to fission; it need not ever be actively removed, the way solid spent fuel is. None is wasted in enrichment, and none is wasted in the reactor. As a result, the economics of nuclear power improve dramatically. Projections show a factor of a hundred gain, possibly several hundred, in energy production over modern-day reactors per unit of unenriched fuel. As a result, very little uranium would be needed at all. One well-known estimate concludes that all human



Unenriched uranium chloride: inexpensive, low-risk fuel for a molten-salt breeder reactor.

it is both created and consumed directly inside the reactor, where it is always dissolved in molten salt. Stealing it would be so wildly impractical as to be effectively impossible.

#### **Pure plutonium**

The Oak Ridge experiments used the FLiBe fluoride salt because it could be integrated well as both coolant and a component of their uranium-tetrafluoride fuel medium. But a chloride-based salt (and fuel) would have important benefits over a fluoride salt. Chloride salt is heavier, which improves the energy distribution of the neutrons that breed plutonium-239 and cause it to fission. It can dissolve more fuel, which means it can also accommodate a greater buildup of fission products (the resulting halves of the "split" atom) without the fuel becoming too dilute. It is lower in viscosity and therefore easier

## RENEWABLE NUCLEAR WOULD BE **EFFECTIVELY INEXHAUSTIBLE**, INHERENTLY SAFE, **LOW-WASTE**, NO-CARBON, **ENRICHMENT-FREE** POWER ON DEMAND. **AND IT'S REALISTIC**.

energy needs could be met for five billion years—until the sun swells up and swallows the earth—without mining any uranium ore whatsoever and instead using only a fraction of the uranium naturally found in seawater.

The MSR would also be vastly safer than current reactors. Meltdowns, for example, would be impossible. The fuel is already molten in normal use, and if it overheated, it would melt a plug and drain into a secure containment vessel. And it is not pressurized, so even if it were released into the environment, it would simply flow out and solidify, not explode. In addition, the public-safety risk of enriched material falling into the hands of terrorists or other enemies would be effectively eliminated. Fissile material is never manufactured, stored, or transported; rather,

to pump through the reactor and other components. And it is safer and more economical to purify. The only real hiccup is the fact that chloride salt hasn't undergone the same extensive testing as fluoride salt. This is where Monreal and Jackson are particularly well positioned to help.

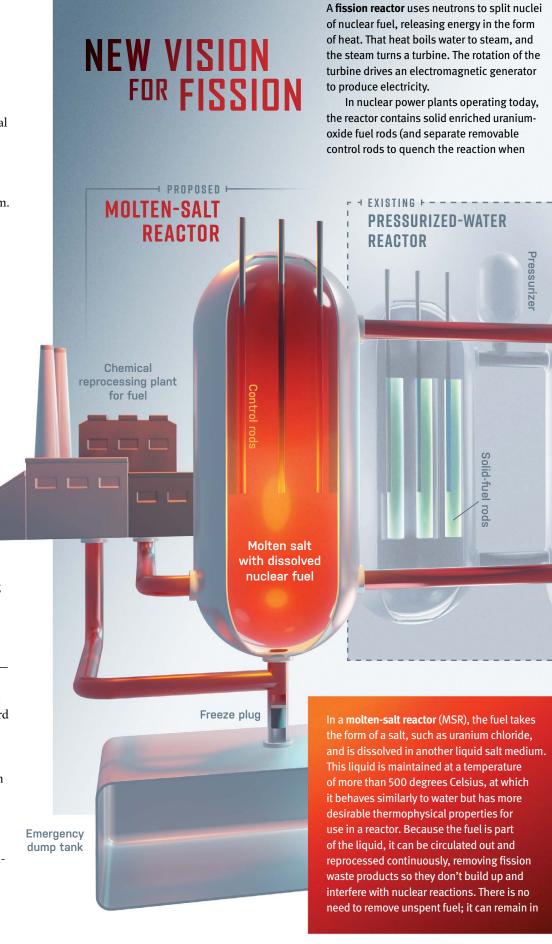
One might wonder why they, as Los Alamos scientists, would be working on a new nuclear-power reactor. While certainly known for nuclear research, Los Alamos's portfolio is weighted toward national security over commercial power. But Monreal and Jackson didn't just leap into commercial power. They started squarely in the middle of the Lab's national security mission space, developing another key molten-salt technology to address a pressing challenge.

"Los Alamos has been tasked to ramp up annual production to 30 plutonium pits—the business end of a nuclear weapon—by 2026," says Jackson. "This is a steep increase over our current capability." The ramped-up production requirements coincide with a national weapons-modernization initiative, and meeting them will not be easy; there is no simple set of knobs to turn that will generate a significant increase in weapons-grade plutonium. The effort will require new workers, new facilities, and above all, a lot of workable plutonium.

Plutonium isn't found in nature; it must be deliberately manufactured from other elements, much like the breeder MSR will do. Some can be recovered directly from decommissioned nuclear weapons, and this is indeed the primary strategy for meeting the pit-production goals. But most of the plutonium available today was produced during the Cold War and purified through a process that leaves it in oxide form: PuO<sub>2</sub>. The trouble is, for a weapon, the oxygen must be removed and the plutonium reprocessed to obtain pure plutonium metal. Doing so by current methods is delicate, painstaking work, requiring rigorous and expensive controls to make it safe. It is also a serious production bottleneck.

Monreal and Jackson have been successfully pioneering a better way—one that will allow them to support the current pit-production challenge and expand capabilities going forward to take advantage of the larger supply of plutonium that exists in oxide form.

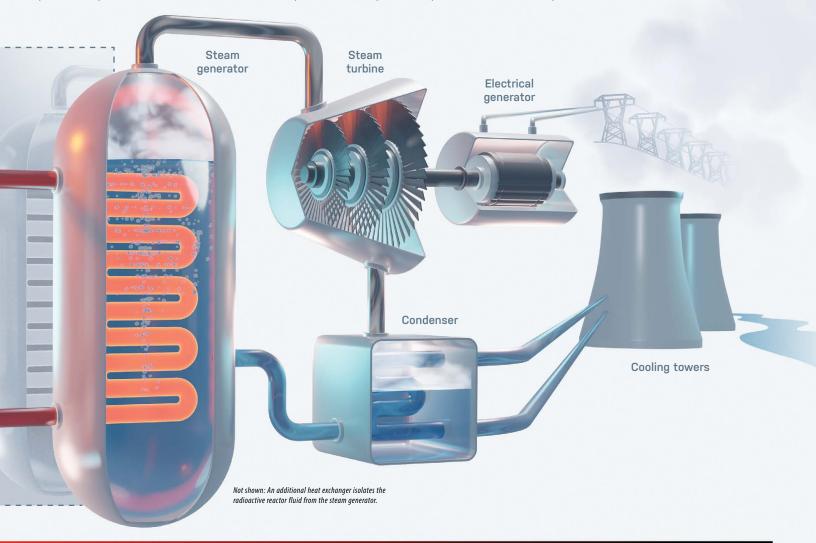
Due to the earth's reactive oxygen atmosphere, metals in nature are often bound to oxygen. In some cases, removing the oxygen (a process referred to as "reduction") can be accomplished with a minimal-resource, low-waste catalytic process taking place in an electrochemical cell, similar to a battery, but filled with—you guessed it—molten salt.



needed) submerged in a fluid moderator medium, usually water, which slows down neutrons to help them induce more fissions and carries heat to the steam generator. The fuel rods become progressively less effective over time and must be replaced at regular intervals despite still containing fissile (fission power-enabling) uranium-235. The regular insertion of fresh fuel rods requires the regular enrichment of nuclear

fuel—an energy-intensive, costly, and wasteful process—that increases the uranium-235 content from about 1 percent in naturally occurring uranium to about 4 percent. The other 96 percent remains non-fissile uranium-238, which ultimately becomes nuclear waste.

The reactor vessel is pressurized, and in the event of an overheating accident, such as a natural disaster that overwhelms the system's many redundant safeguards and prevents cooling water from circulating through the reactor, temperatures and pressures can rise beyond their design tolerances. Solid fuel can undergo meltdown, leading to more fission, greater overheating, and either a controlled venting or an uncontrolled explosion that disperses radioactive gases widely into the atmosphere and possibly contaminates nearby lakes or rivers that have been tapped for cooling water.



place until fission is induced, so little to none is wasted. The energy output per fuel input may be hundreds of times greater than that in existing nuclear reactors.

Molten salt is a poor moderator. An additional moderator can be added, or the neutrons can remain unmoderated, zipping about at high speed. This reduces the fission rate but increases the rate at which non-fissile uranium-238, which makes up more than 99 percent of naturally occurring uranium on Earth, is converted to fissile plutonium-239. Such a molten-salt fast reactor (MSFR) is also

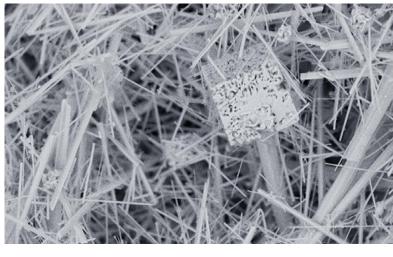
known as a breeder reactor, in that it produces, or breeds, fissile fuel faster than it consumes that fuel. In this sense, an MSFR borders on being a renewable-energy system. In addition, it runs on a small amount of uranium-238, eliminating the need for uranium enrichment entirely, and it can consume certain dangerous isotopes during normal operation, thereby downgrading nuclear waste. Furthermore, because the fissile fuel is produced and consumed entirely inside the reactor, it cannot be stolen, whereas enriched solid fuel can be stolen at the enrichment facility, the power plant, or in transit between the two.

A nuclear accident in any type of MSR, or even deliberate sabotage, would not produce a meltdown because the fuel is molten already. In the event of overheating, the molten salt, which is not pressurized, melts a plug and drains harmlessly into an underground containment vessel. This occurs naturally without any operator actions, mechanical systems, or computer controls, so there is no danger of radioactive release. And even if an earthquake or similar low-probability disaster were to produce a release, the molten salt would solidify in place rather than disperse into the environment.

The process had already been discovered in academia and successfully employed and scaled up for the production of titanium. Monreal and Jackson suspected they could make it work for plutonium.

#### **Essential electrolytes**

Of course, even at Los Alamos, one of the few places capable of working with plutonium, one does not simply grab plutonium oxide off the shelf and begin experimenting with it. So Monreal and Jackson, joined by Los Alamos engineering colleague Kirk Weisbrod, set up a research plan to test surrogate metal oxides first. Weisbrod was instrumental in designing and building a suitable electrochemical cell—a molten salt-filled vessel with two electrodes and a current-carrying external circuit in between. First, they tested it, successfully, with tin oxide. Then they tried cerium oxide because cerium is one step closer to matching the electrochemical properties of plutonium. This was also successful, but there were complications because of several stable, partially reduced cerium compounds, such as CeOCl and Ce<sub>2</sub>O<sub>3</sub>, to pass through along the way from CeO<sub>2</sub> to cerium metal. The final step, currently underway in a collaboration with other interested laboratories, is testing the process with plutonium oxide. This should actually be easier than cerium oxide, since the reduction mechanism for plutonium oxide mitigates problematic partially reduced compounds. Additionally, plutonium oxide is conductive at higher temperatures, therefore improving overall efficiency.



Scanning electron microscope image of CaSnO<sub>3</sub> (a calcium-tin-oxide, or calcium stannate) formed on the anode during a molten-salt electrochemical experiment intended to strip the oxygen from tin oxide in preparation for a similar experiment with plutonium oxide. These crystals—both the needles and the cubes come from this same compound—are electrically insulating; as they built up, the electrochemical cell lost functionality. The problem can be solved with a different anode material or a different metal oxide. CREDIT: Scott Parker/LANL

bottom. Then chlorine gas is added in order to convert the excess calcium and the calcium-oxide byproduct (containing the oxygen removed from the plutonium) back to calcium chloride; upon chlorination, the oxygen is allowed to escape.

It may sound straightforward, but there is a three-fold difficulty. First, both added ingredients, calcium and chlorine, are hazardous materials, requiring safety precautions and monitoring by trained personnel. Pure calcium is explosively reactive in water, and chlorine gas is poisonous. Second, even though the process has been greatly optimized, it still requires these two separate steps—adding calcium to remove the oxygen, adding chlorine to restore the salt—which must be repeated four times to obtain the plutonium metal in a typical batch. This is quite time-consuming. Third, and also time-consuming, is a separate purification step needed at the end. Contaminants are removed from the plutonium metal through a process called electrorefining, which

# ISOLATING PLUTONIUM METAL BY CURRENT METHODS IS COMPLICATED WORK— AND A SERIOUS PRODUCTION BOTTLENECK.

Molten salt already plays a role in Los Alamos's existing process for reducing plutonium oxide to plutonium metal, but that is a purely chemical process, not an electrolytic one. In this existing chemical process, the  $PuO_2$  and granulated calcium are mixed into molten calcium chloride salt, which is used here as both a solvent and a heat sink. The calcium displaces the oxygen and liberates plutonium, which settles to the

involves setting up a current in a separate electrochemical cell. When all is said and done, the molten salt is no longer usable and is disposed of as radioactive waste.

In the new process, the same calcium chloride salt is used, this time as an electrochemical conductor. The negatively charged electrode, or cathode, catalyzes the key swap:

separating oxygen from the plutonium and attaching it instead to calcium ions in the salt. The calcium oxide thus produced then finds its way to the positively charged anode, where the oxygen is stripped from the calcium oxide and removed, restoring calcium ions to the salt. That reaction produces free electrons, which are then carried as a current along an external circuit to the cathode, restoring the negative charge there to continue the process. Neither granulated calcium nor chlorine gas is needed, removing the hazards so that the process may run unattended, and the salt is restored for future reuse, rather than discarded. Furthermore,

Monreal and Jackson are hopeful that since both the reduction and purification processes are electrochemical in nature, they can be combined in the same electrochemical cell. All in all, the total processing time can be significantly decreased.

#### Salt substitute

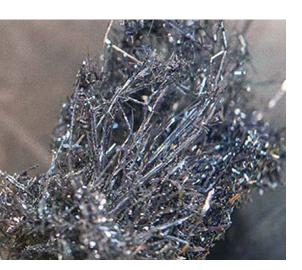
Now that Jackson and Monreal have built and tested their electrochemical cell for plutonium reduction and purification, it is available as a testbed for other experiments—in particular, for research that might facilitate the adoption of essentially renewable nuclear power. Ultimate success would offer effectively inexhaustible, inherently safe, low-waste, no-carbon, enrichmentfree, ample power on demand, whether it's sunny or windy or neither. And it's realistic.

What does MSR-based nuclear power need to become a reality? "A lot of things, but none of them is prohibitive," says Monreal. "We just need to further develop our facility for working with these molten chlorides. We need to continue to measure them, model them, control them. We need to develop the industry to go along with the reactor."

The industry Monreal mentions will be one of sensors, systems, software, and safeguards. None of this is easy; even just the sensors—which will be needed to monitor the fluid properties, contaminant levels, and corrosivity of the salt solution in the reactor—do not exist. How do you reliably and continuously measure the properties of molten lava? Even for something as simple as its volume, which is a key component of density, it is not immediately obvious how to measure it inside an opaque, sealed furnace, using only components that can withstand the heat.

"I think we can design a new kind of dilatometer to handle that," says Jackson, referring to an instrument that will sense the expansion of the salt as it is heated. "But that's just volume. Other measurements will be harder."

One of these harder measurements is fluid viscosity—say, at different temperatures or with different levels of actinide fuels or fission products mixed in. It is an important parameter to understand for MSRs, but the most direct method—dropping spheres of different densities through the medium and recording their fall time—requires seeing inside the furnace. Jackson and Monreal are using an advanced Los Alamos capability, neutron imaging, to see through the furnace instead. The research



Using a modified configuration of the electrochemical cell, these high-purity uranium dendrites were produced from impure uranium metal in a molten-salt electrolyte. This purification step comes after the conversion from metal oxide to metal. CREDIT: Matt Jackson/LANL

plan is to conduct this experiment repeatedly in a laboratory setting—experimentation that's already underway and producing results—in order to tabulate the range of viscosity and density values for the salt at different temperatures and actinide concentrations. Such data would then feed into new simulation software for predicting the molten material's behavior as conditions are changed, for plant operation or optimization purposes.

Because the medium will support nuclear reactions, it will be creating different elements as it runs—breeding plutonium, splitting into fission products, absorbing neutrons, radioactively decaying from one element or isotope to another—and any or all of these changes could result in markedly different properties, which is why Jackson and Monreal are experimenting with different compositions and concentrations. Some might alter

### WHEN VERY LITTLE FUEL IS WASTED, VERY LITTLE FUEL IS NEEDED.

the overall heat capacity or conductivity. Some might spawn the production of corrosive agents. Some might require specific types of chemical analysis and processing alongside the reactor. Some might imply a tradeoff: do X and you'll get better Y but worse Z. To make MSR-based renewable nuclear power a reality, all of this must be known.

And all of it will be.

The United States currently gets about 20 percent of its electricity from 97 nuclear reactors with an average age of nearly 40 years. All of them run on enriched solid fuel, none is a breeder, and none is anywhere near as safe, efficient, or environmentally friendly as an MSR. If it sounds like the time is right for a nuclear renaissance, the experts agree.

"We could see molten-salt reactors ready for commercial construction in the near future," says Monreal, "once we master molten salt." LDRD

—Craig Tyler

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